Assessing Exposures to 1-chloro-4-(trifluoromethyl) Benzene (PCBTF) in U.S. Workplaces

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INTRODUCTION

The chemical compound 1-Chloro-4-(trifluoromethyl) benzene (CAS No: 98-56-6)—also known as PCBTF, Oxsol 100, or Parachlorobenzotrifluoride—was nominated to the National Toxicology Program (NTP) for toxicity and carcinogenicity studies (http://ntp.niehs.nih.gov/ntp/noms/support_docs/pcbtf06-0409.pdf). The nomination was based on the increasing use of PCBTF by industries and consumers, since it was exempted by the Environmental Protection Agency (EPA) as a volatile organic compound in emissions reporting on the basis of not reacting in a manner that would contribute to the formation of tropospheric ozone.[1] Although PCBTF is no longer manufactured in the United States, approximately 29 million lbs. were imported in 2012[2] and used in various applications to replace other chlorinated solvents with known environmental or human health hazards. Those applications include the automotive industry as industry-wide applications in coatings, thinners, and cleaning solvents, and repair and maintenance cleaning and as a consumer product for cosmetic stain removal and aerosol rust prevention.[3]

The toxicity information on PCBTF is available from various resources[4,5] including the NTP website.[6] These studies, however, are limited to short-term toxicity, and chronic inhalation toxicity and carcinogenicity studies are unavailable. There are no Occupational Safety and Health Administration (OSHA) regulations specific to limiting occupational exposures to PCBTF. The National Institute for Occupational Safety and Health (NIOSH) has not established a time-weighted average (TWA) recommended exposure level, and the American Conference of Governmental Industrial Hygienists (ACGIH®) has not established

a TWA-threshold limit value (TLV®) for PCBTF. The Occidental Chemical Corporation, which used to manufacture PCBTF in the United States, established a corporate exposure limit (CEL), which was a TWA limit of 25 ppm (185 mg/m^3) for an 8-hr work-shift. The toxicological basis for setting this limit is not known to us. However, Occidental Chemical Corporation no longer manufactures or imports PCBTF into the United States.

The purpose of this case study is to determine industry-wide occupational inhalation exposures using available industrial hygiene sampling methods. This information can be used to benchmark exposure concentrations that may be applied in future studies of inhalation toxicity in animal models. In addition, side-by-side samples of a pumped (active) and diffusive (passive) sorbent tubes were taken to compare concentration ratios between the active and passive sampling methods.

Workplace Description

Vehicle manufacturing plants—Four vehicle manufacturing plants—helicopter (Plant A), aircraft (Plants B and C), and automobile (Plant C)—were recruited through personal contacts. All manufacturing plants were identified by code for confidentiality. At Plant A, PCBTF was used as a cleaning solvent to remove residual glue after upholstery removal during interior refurbishment. The cleaning work was done manually under a slotted back-draft ventilation hood. PCBTF was used during primer application prior to coating of an airplane at Plants B and C and plastic adhesive promoter application at Plant D. All painters wore airline respirators and applied the PCBTF-containing substances using spray guns under downdraft ventilation. The mixing worker at Plant C combined base (23 L with 0% PCBTF), activator (23 L with 30–60% PCBTF), and thinner (6 L with 60–90% PCBTF) to make primer. The mixing task was done under a canopy hood and the mixer wore a full facepiece air-purifying respirator. The amount of PCBTF per worker used during the specific tasks varied ranging from 0.3 to 18.5 L. Table I shows a summary of workplace description including tasks, PCBTF usage, room ventilation, local exhaust ventilation, respirator type, and the amount of PCBTF used during each task. Detailed information about job tasks and personal protective equipment was described in a supplementary file.

Paint manufacturing plants—Three paint manufacturing plants were recruited via contacting American Coatings Association. Four tasks—pre-batch making, batch making, filling, and miscellaneous—were observed. In the pre-batch making area (Plants E and G), workers transferred PCBTF-containing materials to other containers using either a pumping system or a mechanized pouring system. Containers were partially opened to place a pumping system. No respirator was required for this task at both plants. In the batch-making area (Plants E, F, and G), each batch-maker added various chemicals in a batch container, mixed the chemicals, transferred the chemicals to other containers, and cleaned the emptied batches. The batch-making task was done in a closed system for all plants except for cleaning or partially opened to add or transfer materials. The batch-makers wore no respirators during mixing but wore dust masks (Plants E and G) and half facepiece respirators (Plant F) when manually adding materials. The filling operators (Plants E, F, and G) filled containers with final product from an automated dispenser and placed lids. No respirator was required for the filling task. Other miscellaneous tasks included lab quality

control testing, cleaning, batch adjusting, color mixing, and pilot working. The workplaces for all tasks were controlled by general ventilation in addition to any local exhaust ventilation systems. Table II shows a summary of workplace description and detailed information for each task was described in a supplementary file.

METHODS

Sample Monitoring

At the four vehicle manufacturing plants, 28 personal and 8 area sample pairs were collected using actively pumped coconut-shell charcoal tubes (SKC 226-01, SKC Inc., Eighty Four, PA) and diffusive charcoal badges (SKC 575-001, SKC Inc.). The former represents an active sampling method (i.e., drawing air throughout the media using a pump) and the latter represents a passive sampling method (i.e., air intake by chemical diffusion). All workers sampled at the vehicle manufacturing plants handled the PCBTF-containing materials.

At the three paint manufacturing plants, 64 personal and 26 area sample pairs were collected. Participants were workers who handled PCBTF and workers who did not but were in close proximity to the workers handling PCBTF. The sample size and sampling time for each task are listed in Table III. The sampling times ranged from 15 to 407 min for the vehicle manufacturing plants and 70 to 535 min for the paint manufacturing plants. Two types of sampling pumps, Pocket Pump (SKC Inc.) and Gilian LFS-113 (Sensidyne, Clearwater, FL), were used at sampling flow rates between 20 and 200 ml/min for the active sampling method. The sampling flow rates were adjusted based on anticipated concentrations, previously collected from similar workplaces. Each pump was calibrated before and after sample collection with a DryCal DC-Lite device (BIOS International Corporation, Butler, NJ) to assure the difference between pre- and post-sampling flow rates was within ±5%. The position of passive and active samplers for the personal sampling method was randomized to minimize bias from workers' handiness (i.e., not always on the left or right of worker's collar). All field surveys were performed between 2010 and 2012.

All active and passive samples were analyzed with gas chromatography/flame ionization detector according to the *NIOSH Manual of Analytical Methods* (NMAM) 1026[7] by the NIOSH contract laboratory. The NIOSH method has suggested a maximum of 25 ppm for a 10 L air sample with a working range between 0.024 and 9.15 ppm (0.178 to 67.8 mg/m^3). Yost and Harper[8] tested passive badges at various loadings in a standard atmosphere chamber in which the test concentrations of the standard atmosphere were confirmed by means of coconut charcoal tubes for time period up to 8 hr. Those loadings were 0.012 mg (0.01×CEL), 0.123 mg (0.1×CEL), 0.505 mg (0.5×CEL), 1.10 mg (1.0×CEL), and 2.09 mg (2.0×CEL). Yost and Harper[8] showed charcoal tubes and passive badges to have a large capacity covering up to 2 times the CEL and the maximum concentration suggested by the NIOSH method.

The mass concentrations of passive badges were calculated using the average sampling rate of 11.8 ml/min.[8] From each sampling site, 1–10 field blank samples were collected. In this study, sample results were not adjusted by field blank samples because almost all field blank samples (96% of 56 field blank samples) showed non-detectable masses. The limit of

detection was 0.1 – 0.7 *μ*g for both diffusive badge and charcoal tube. The limit of quantitation ranged between 0.5 – 2.5 *μ*g for the diffusive charcoal badge and 0.5 – 3.4 *μ*g for the charcoal tube. Three sample pairs showing at least one of each pair resulted in less than the limit of detection were excluded. None of the samples except for the three sample pairs showed less than the limit of quantitation.

RESULTS

Exposure Assessment

Table III shows air sampling results using the active sampling method at the vehicle and paint manufacturing plants. Overall, the geometric mean of personal exposures (2.1 ppm) at the vehicle manufacturing plants was 3 times higher than that (0.7 ppm) at the paint manufacturing plants, while the geometric means of area exposures were similar for both types of plants. For the combined results of personal and area samples, the geometric mean concentration at the vehicle manufacturing plants was higher (about 2.5 times) than at the paint manufacturing plants. The comparison of log-transformed exposures between the vehicle and the paint manufacturing plants resulted in statistically significant difference (p-value < 0.0001). None of the samples exceeded even one-half the Occidental Chemical Corporation in-house CEL of 25 ppm.

Vehicle Manufacturing Plants

For the vehicle manufacturing plants, all individual measurements per task were less than 10 ppm except for one measurement (12.2 ppm) from an interior refurbishment worker at Plant A (Figure 1). Although the amount of PCBTF handled during the mixing was larger than the other two tasks (interior refurbishment and painting tasks), the differences of geometric mean exposure concentrations between tasks were not substantial. Note that the mixing task could not be compared to other tasks because only one measurement was obtained. For the primer painting task, Plant C showed higher concentrations than Plant B shown in Figure 1. The geometric means of area and personal exposures for all tasks and areas were less than 3 ppm (Table III). The geometric standard deviation (GSD) per task was high (i.e., > geometric mean per task), indicating a wide spread of exposure measurements.

Paint Manufacturing Plants

For the paint manufacturing plants, all collected exposure measurements were lower than 8 ppm. Note that personal exposure measurements of lab quality worker (n = 1) at Plant E, cleaner (n = 3) and batch adjuster (n = 1) at Plant F, and mixer (n = 3) and pilot worker (n = 3) at Plant G were merged into the category "miscellaneous" due to small sample sizes (3). Overall, although slight differences in geometric mean concentrations between tasks were observed, the magnitudes of differences were negligible. The variations of exposure measurements were high for most tasks (i.e., high GSD). It was also noted that the exposures measured at Plant E were higher than at the other two plants when personal and area exposures of pre-batch making and batch-making tasks were compared (Figure 2). Unlike the vehicle manufacturing plants, area geometric mean exposures were higher than the personal exposures when comparison was made by task. At Plant G, workers' exposures

who did not handle the PCBTF compound were about 20–55% lower than those exposures who handled PCBTF during full-shift.

Comparison of Active and Passive Sampling Methods

Figure 3 presents side-by-side sample results (i.e., active versus passive) for the vehicle and paint manufacturing plants. Comparison by job task was not performed due to insufficient sample sizes per job task. The slope developed from linear regression method of log-transformed data was 0.929 with adjusted $R^2 = 0.606$ for the vehicle manufacturing plants and 1.012 with adjusted $R^2 = 0.788$ for the paint manufacturing plants. The slope was 0.990 with adjusted $R^2 = 0.773$ for the combined data. All p-values for testing H_o: slope $(\beta) = 1$ were > 0.05 indicating statistically no significant differences of concentrations between active and passive samples.

As shown in Table IV, the median of concentration ratios (passive/active) ranged from 0.9 to 1.2 for the personal and area samples. The geometric mean of concentration ratios was 1.0 regardless of the type of workplace and sampling method (e.g., personal versus area). For the paint manufacturing plants, more variation of the concentration ratios (passive/active) was observed from the area samples compared to the personal samples, while the area samples showed less variation than the personal samples for the vehicle manufacturing plants. A strong correlation was observed for all pairs of samples (all correlation coefficients 0.763), indicating statistical significance (all p-values < 0.0001 except for area samples at vehicle manufacturers (p-value $= 0.0014$)). No overall statistical differences were observed from the comparison of exposure measurements between the active and passive samples (all p-values > 0.05) regardless of worksite type. Additionally, the separation of personal and area exposures did not yield statistically different results (all p-values > 0.05).

DISCUSSION

Exposure Assessment

The PCBTF exposure measurements (personal and area combined) were 0.1–12.2 ppm (geometric mean 2.0 ppm, GSD 2.9 ppm) for the vehicle manufacturing plants and 0.1–7.7 ppm (geometric mean 0.8 ppm, GSD 3.1 ppm) for the paint manufacturing plants. All individual measurements represented task-specific exposures but only some tasks extended close to a full 8-hour shift. Regardless of sampling time period, all measurements were considerably lower than a previous manufacturer's in-house CEL of 25 ppm. The geometric mean exposure was only 8% of the CEL for the vehicle manufacturing plants and 3.2% of the CEL for the paint manufacturing plants. If the CEL is selected as an occupational exposure limit (OEL) value, it is very likely that none of tasks involving handling PCBTF measured in this study would result in the employee being overexposed. Similarly, a low exposure range was also observed in a survey performed at the Occidental Chemical Corporation. They monitored personal exposures from operators working in a cold cleaning machine containing PCBTF to remove grease and dirt from metal parts. The amount of PCBTF used in the machine was about 300 L and a solid metal lid covered the opened top. The observed personal exposures during a 3-week period ranged from 0.008 to 2.6 ppm

(average 0.7 ppm, SD 0.9 ppm) for the operators working at the machine approximately 20 min per full-shift (Personal communication).[9]

The PCBTF inhalation exposures were determined by the combined effect of the PCBTF characteristics, amount handled during tasks, surface area of solvent exposed to air, handling method (e.g., spraying or manual paint brush application), and control strategy during the handling of the substance. For example, although large amounts of PCBTF were handled during the batch-making task (up to 3200 L), the partial opening of the batch for adding/ transferring materials (i.e., enclosed system) did not result in high exposure to workers (geometric mean of personal exposures for all paint manufacturers = 0.8 ppm). Similarly, the geometric mean exposure of the pilot worker was 0.5 ppm when spraying and testing the PCBTF-containing material under a local-exhaust fume hood. If the pilot work was done without the presence of a local-exhaust hood, the PCBTF exposure would likely have been higher than 0.5 ppm. Although it would be difficult to determine which exposure determinants were more important than others without sensitivity analysis, one of the main reasons for the low exposure ranges is likely to be the relatively low vapor pressure of PCBTF (5.3 mmHg at 20°C) compared to other chemicals such as acetone (180 mmHg at 20°C).

Painter exposures measured at Plant C and exposures of pre-batch maker and batch maker at Plant E were higher than the same tasks of other plants. The different workplace conditions such as ventilation method, room sizes, and tools used to perform the tasks might cause such differences. However, another factor could be the sampling times in comparing to the time of specific use of PCBTF-containing materials. For example, the sampling time of painter at Plant B (85 and 111 min) was about 3 times longer than that at Plant C (32 and 37 min). Communication with the industrial hygienist who collected samples at Plant B confirmed that the painter exposures included the task handling materials with and without PCBTF. Inclusions of sampling time other than the painting task such as painting preparation prior to and cleaning after the painting task would result in lower concentration compared to the painting task handling the PCBTF only (geometric mean: 1.4 ppm at Plant B versus 9.5 ppm at Plant C). In the present study, the sampling time and task time are not necessarily equivalent due to the nature of task performance.

Personal and area measurements showed relative differences between the two worksite types. For the vehicle manufacturing plants, the geometric mean of personal exposure measurements was about 1.3 times higher than that of area measurements, while the geometric mean of area measurements for the paint manufacturing plants was 1.8 times higher than that of personal exposure measurements. This difference was very likely due to the distance of the sampler's location from the source location. Although the geometric mean of area measurements was higher than that of personal exposure measurements for the paint manufacturing plants, this would not change the general conclusions of this study.

This case study was limited to the range of room temperature from 13°C to 27°C. Tasks at temperatures higher than the temperature range in this study might reveal different findings. Also, most samples were collected less than a full-shift (i.e., 8-hr TWA) and do not necessarily reflect 8-hr sampling.

Comparison of Passive and Active Exposures

No statistical differences between active and passive sample measurements were determined. For the vehicle manufacturing plants, the variation of the ratios of passive/active concentrations was higher for the personal exposure measurements than for the area measurements. On the other hand, for the paint manufacturing plants, a higher variation was observed from the ratios of area measurements than the ratios of personal exposure measurements. Higher variation between personal sampling methods may be the result of a worker's movement and position of the sampler against the source (e.g., one sampler closer to the source than the other one), whereas the area sample pairs were fixed adjacent to each other. There is no clear explanation for the higher variation of the area sampling methods for the paint manufacturers. The findings of this study support the use of a passive sampler as an alternative to the active sampler to assess exposures to PCBTF in the vehicle and paint manufacturing plants.

CONCLUSION

Industry-wide occupational exposures to PCBTF were determined by assessing workers' exposure from personal and area samples in various tasks in vehicle and paint manufacturing plants. Tasks monitored in this study were interior refurbishment worker, painter, and mixer from four vehicle manufacturing plants and pre-batch maker, batch maker, filler, and miscellaneous (including mixer, cleaner, pilot worker, lab quality worker, and batch adjuster) from three paint manufacturing plants. None of the individual measurements exceeded 13 ppm, considerably lower than a previously proposed in-house CEL of 25 ppm. The range of occupational exposures in this study would provide guidance on animal toxicity research conducted or supported by the NTP and provide human exposure data needed for policy-making. However, measurements were not made in other industries where exposures could be higher than those observed here, such as in autobody repair and refinishing shops.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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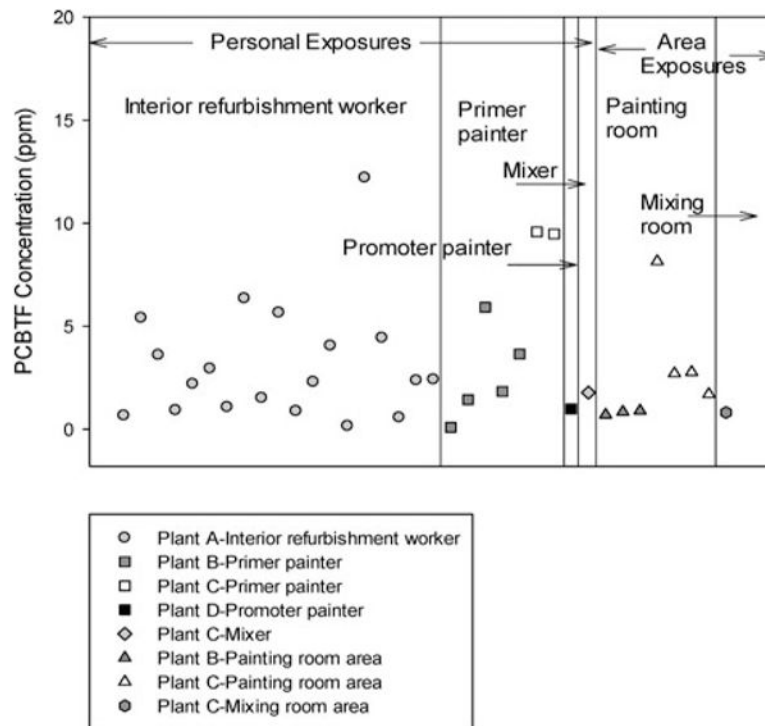
FIGURE 1.
Individual exposure measurements by plant per job task (Vehicle manufacturers).
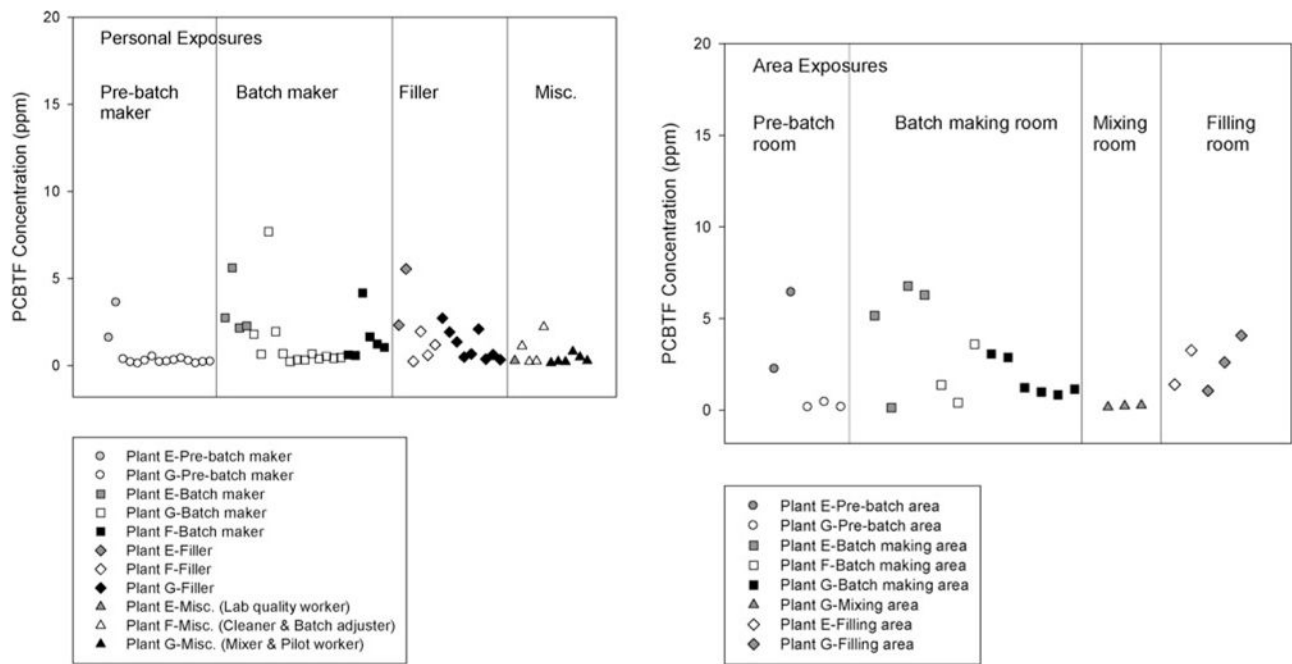
FIGURE 2.
Individual exposure measurements by plant per job task (paint manufacturers).
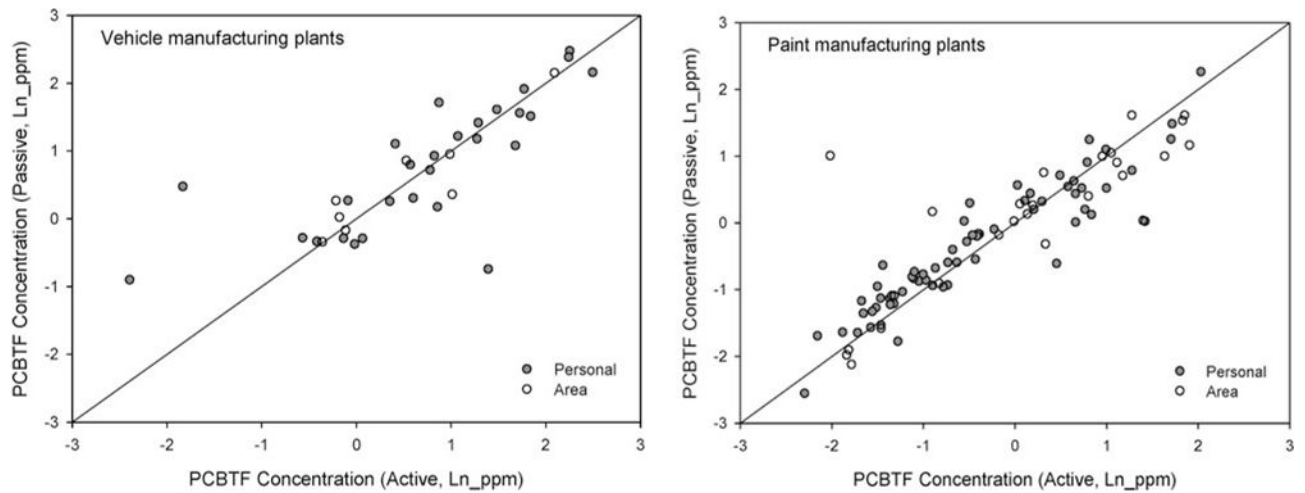
FIGURE 3.
Log-transformed PCBTF concentrations between the pairs of samples. The diagonal line represents 1:1 relationship. (Vehicle: Ln_Passive = 0.011 + 0.929*Ln_Active with adjusted R^2 = 0.606, Paint: Ln_Passive = −0.022 + 1.012*Ln_Active with adjusted R^2 = 0.788).

TABLE I

Summary of Workplace Description (Vehicle Manufacturing Plants)

Job title	Interior refurbishment worker	Primer painter	Mixer	Promoter painter
Plant	A-Helicopter industry	B and C-Aircraft industry	C-Aircraft industry	D-Automobile industry
PCBTF usage	Cleaning solvent (manually)	Primer	Primer mixing	Plastic adhesion promoter
Room ventilation	General ventilation	General ventilation[A]	General and natural ventilation	General ventilation
Local exhaust ventilation	Slotted back-draft hood	Downdraft ventilation	Canopy hood	Downdraft ventilation booth
Respirator	None	Hood airline respirator	Full facepiece air-purifying respirator	Hood airline respirator
PCBTF amount/worker (liter)	0.3 – 1.0	3.0 – 5.0	< 1.0	10.0 – 18.5

[A]The room was controlled by general ventilation while no painting work was performed.

TABLE II

Summary of Workplace Description (Paint Manufacturing Plants)

Job title	Pre-batch maker	Batch-maker	Filler	Miscellaneous[A]
Plant	E and G-Paint industry	E, F, and G-Paint industry	E, F, and G-Paint industry	E, F, and G-Paint industry
PCBTF usage	Transferring of chemicals to other containers	Mixing/adding materials; cleaning empty containers	Filling paint materials	Various tasks including lab quality, cleaning, batch adjusting, color mixing, and pilot working
Room ventilation	General ventilation[B]	General ventilation[B]	General ventilation	General ventilation
Local exhaust ventilation	Slotted back-draft hood (Plant E); None (Plant G)	Mixing in an enclosed system; local exhaust flexible duct while adding materials manually	4-inch flexible duct near fill-heads (Plants E and F); None (Plant G)[C]	Cleaner (Plant F) - slotted back-draft hood; Batch adjuster and Mixer (Plants F and G)- flexible local exhaust duct; Pilot worker (Plant G)-Local exhaust ventilation hood
Respirator	None	None during mixing; dust mask (Plants E and G) and half facepiece respirator with dual cartridges for VOC and particulates (Plant F) for manually adding materials	None	None for the lab quality worker (Plant E), cleaner (Plant F), and pilot worker (Plant G); half facepiece respirator with dual cartridges for VOC and particulates for the batch adjuster (Plant F); dust mask for the color mixer (Plant G).
PCBTF amount/worker (liter)	NO,[D] $0^E - 1500$	NO,[D] $0^E - 3200$	NO,[D] $0^E - 4500$	NO[D]

[A]Miscellaneous includes those tasks which sample sizes were 3 (including lab quality worker at Plant E, cleaner and batch adjuster at Plant F, and mixer and pilot worker at Plant G).

[B]Plant G also had natural ventilation by opening garage doors near the task area.

[C]The three sides of the automatic dispenser at Plant G were covered with acrylic sheet.

[D]NO = Not Obtained. The PCBTF amount per worker was not obtained at some workplace.

[E]Although a few workers per task did not handle PCBTF during the field survey, we collected samples because they worked next to other workers handling the PCBTF.

TABLE III

Air Sampling Results Using Active Sampling Method

	Task	N[A]	Sampling time (min)	TWA Concentration (ppm)				
				Min	Max	Mean	Geo-mean[B]	GSD[C]
Vehicle manufacturers-all plants	Interior refurbishment	19	92–407	0.2	12.2	3.1	2.1	2.7
	Painting primer	7	32–111	0.1	9.6	4.6	2.4	4.5
	Painting promoter	1	15	N/A	N/A	1.0	N/A	NC
	Mixing	1	78	N/A	N/A	1.8	N/A	NC
	Painting room (area)	7	43–111	0.7	8.2	2.5	1.8	2.3
	Mixing room (area)	1	77	N/A	N/A	0.8	N/A	NC
	All personal	28	—	0.1	12.2	3.4	2.1	3.1
	All area	8	—	0.7	8.2	2.3	1.6	2.2
	All (personal+area)[D]	36	—	0.1	12.2	3.1	2.0	2.9
Paint manufacturers-all plants	Pre-batch making	15	70–449	0.1	3.6	0.6	0.3	2.5
	Batch making	23	163–535	0.2	7.7	1.7	1.0	2.6
	Filling	15	185–479	0.2	5.5	1.5	1.0	2.4
	Miscellaneous (personal)[E]	11	190–483	0.2	2.2	0.6	0.4	2.2
	Pre-batch room (area)	5	70–469	0.2	6.4	1.9	0.7	4.3
	Batch making room(area)	13	150–527	0.1	6.8	2.6	1.6	3.0
	Mixing room (area)	3	443–482	0.2	0.3	0.2	0.2	1.2
	Filling room (area)	5	212–468	1.1	4.1	2.5	2.2	1.7
	All personal	64	—	0.1	7.7	1.2	0.7	2.9
	All area	26	—	0.1	6.8	2.2	1.2	3.5
	All (personal+area)[D]	90	—	0.1	7.7	1.5	0.8	3.1

Notes: N/A = Not Applicable; NC = Not Calculated. Geometric standard deviation (GSD) is not calculated due to a small sample size.

[A] Number of samples collected during the field survey.

[B] Geomean = Geometric mean.

[C] GSD = Geometric standard deviation.

[D] Combined results of personal and area sample concentration (ppm).

*E*Miscellaneous includes those tasks which sample sizes were 3 (including lab quality worker at Plant E, cleaner and batch adjuster at Plant F, and mixer and pilot worker at Plant G).

TABLE IV

Statistical Analysis Results Between Pair of the Samples

Worksite	Sample Method	Median of concentration ratio (Passive/Active)	Coefficient of Variation (CV)[A]	Pearson Correlation Coefficient (p-value)[B]	P-value of Proc Mixed (Concentration Estimates)[C]
Vehicle manufacturers	Personal	1.1	1.3	0.763	0.944 (A: 1.99 ≅ P: 2.03)
	Area	1.0	0.3	0.915	0.853 (A: 1.40 ≅ P:1.44)
	All	1.1	1.2	0.786	0.887 (A: 1.67 ≅ P: 1.62)
Paint manufacturers	Personal	1.2	0.3	0.929	0.595 (A: 0.82 ≅ P: 0.80)
	Area	0.9	2.2	0.803	0.473 (A: 1.35 ≅ P: 1.54)
	All	1.1	1.6	0.889	0.678 (A: 0.88 ≅ P: 0.90)
Vehicle + Paint manufacturers	Personal	1.1	0.9	0.901	0.459 (A: 1.17 ≅ P: 1.23)
	Area	1.0	2.2	0.819	0.999 (A: 1.37 ≅ P: 1.37)
	All	1.1	1.5	0.880	0.570 (A: 1.24 ≅ P: 1.28)

[A] Coefficient of variation (CV) = standard deviation of ratios (passive/active) divided by average ratio of concentration (passive/active).

[B] Pearson correlation coefficient with log-transformed concentrations. All p-values for the comparison were < 0.0001 except for area samples at vehicle manufactures (p-value = 0.0014).

[C] Concentration estimates of active samplers (A) and passive samplers (P).